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Mazurek, P.; Hvilsted, S.; Skov, A. L.

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Green silicone elastomer obtained from a counterintuitively stable mixture of glycerol and PDMS

P. Mazurek

S. Hvilsted

A.L. Skov*

al@kt.dtu.dk

Danish Polymer Centre, Department of Chemical and Biochemical Engineering, Building 227, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

*Corresponding author.

Abstract

A green and cheap silicone-based elastomer has been developed. Through the simple mixing-in of biodiesel-originating glycerol into commercially available polydimethylsiloxane (PDMS) pre-polymer, a glycerol-in-PDMS emulsion was produced. This counterintuitively stable mixture became a basis for obtaining elastomeric composites with uniformly distributed glycerol droplets. Various compositions, containing from 0 to 140 parts of glycerol per 100 parts of PDMS by weight, were prepared and investigated in terms of ATR-FTIR, broadband dielectric spectroscopy, mechanical properties as well as optical and scanning electron microscopy. The materials were proven additionally to exhibit a strong affinity to water, which was investigated by simple water absorption tests. Incorporating glycerol into PDMS decreased the Young's modulus of the composites yet the ultimate strain of the elastomer was not compromised, even in the presence of very high loadings. The conducted experiments highlight the great potential of this new type of elastomer and reveal some possible applications.

Keywords: PDMS; Silicone; Glycerol; Filler; Emulsion; Elastomer; Green; Eco; Absorption; Release

1 Introduction

Non-toxic, energy saving, biodegradable, made from recycled materials or renewable resources, made from industrial or agroforestry residues – the qualification list for a polymer to be regarded as “green” is very long. Thus, fulfilling more than one of them generally leads to multiple issues in material preparation procedures, and it usually also compromises the material's ultimate properties [1]. Research on eco-friendly plastics has become one of the main targets of modern industry [2–5], and the prospect of an oil shortage motivates the material industry to look into innovative sources. Ideal alternatives to traditionally synthesised monomers seem to be substrates of natural origin; therefore, extensive investigations into the usefulness of bio-based monomers, as well as natural fillers (e.g. fibres), have been conducted successfully [6–10]. Nonetheless, not many reports have been presented on green elastomers. The significant extensibility of these materials is a very unique property that requires an uncompromised balance of physical and chemical properties in their structure, and so the task of preparing commercially attractive elastomers is a highly complicated procedure. Nevertheless studies exist representing successful attempts at using bio-based substrates and incorporating them into elastomers. An interesting example is the work of Brook et al., who employed softwood lignin, acting as a crosslinker, and at the same time as a reinforcing agent in silicone elastomers [11]. Furthermore, in other approaches the biodegradability of various elastomers has extensively been investigated and finally improved [12–15]. This indicates that there is a need for developing green elastomers, although the preparation procedures still require further optimisation.

It is common knowledge that glycerol, as a residue of biodiesel production, offers great potential in the preparation of a new wave of green polymers. Considering the fact that biodiesel production in European Union increased fourfold between 2004 and 2008, and that 100 kg of glycerol is obtained from 1000 kg of biodiesel, it is believed that more attention should be afforded to research on glycerol-based polymers [16]. Thus, so far, multiple research groups have investigated the usefulness of this compound in organic chemistry and material science [17,18]. One common example of applying glycerol in polymer science is using it as one of the monomers in polymer synthesis [13,19,20]. Introducing glycerol into polymers as one of the substrates brings these materials closer to the definition of “green” polymers, although the amounts of glycerol used in such cases are usually insignificant compared to other reactants, whilst in addition the resulting polymers become significantly more expensive.

Adding solid fillers to elastomeric or non-elastomeric polymers is usually intended to improve material properties or decrease the overall price of the product. In either case, adding fillers significantly influences material features over time, i.e. product reliability is reduced [21]. For example, in the case of polydimethylsiloxane, incorporating silica particles is a vital process that allows for obtaining highly stretchable materials with good ultimate properties. On the other hand, incorporating rather inexpensive fillers, such as wood-dust, is a common procedure for decreasing a product's price [22]. In our approach we present an innovative way of identifying the introduction of fillers into elastomers. We investigated the

possibility of using glycerol as a liquid filler of a polar nature incorporated into a commercially available, highly non-polar, silicone elastomer. We found that incorporating glycerol into PDMS does not compromise the ultimate strain of the material, thereby making it much cheaper compared to non-glycerol PDMS. Samples with different amounts of glycerol were prepared and extensively investigated in terms of their morphology and mechanical properties. The conducted experiments revealed and finally proved the great potential of this new type of green and cheap elastomer.

2 Experimental

2.1 Materials

A two-component silica-filled Sylgard 184 silicone kit (S184) was purchased from Dow Corning US. Glycerol (food grade), as a byproduct of biodiesel production, was provided by Emmelev A/S Denmark and was used as received, avoiding excessively long contact with the air.

2.2 Methods

A dual asymmetric centrifuge SpeedMixer DAC 150 FVZ-K was used for mixing all of the compounds. A Leica DM LB optical microscope was applied for investigating glycerol in silicone emulsion morphology. Viscosities of the investigated pre-polymer formulations were determined with the help of an ARES AR2000ex rheometer from TA Instruments using a conical concentric cylinder geometry at a steady state flow measurement. Data points used for comparison of viscosities were obtained from shear rate of 0.1 1/s. A FEI Inspect S scanning electron microscope was used to obtain images of cured specimens' cross-sections. Samples were gold-sputtered prior to testing, in order to obtain higher quality images. ATR-FTIR spectra of composite cross-sections were obtained with the help of a Nicolet iS50 FT-IR spectrometer, while tensile tests were performed on an Instron 4301 universal tester at room temperature at a strain rate of 500 mm/min – as stipulated by ASTM D412-C standards. Sample conductivity was investigated with a Novocontrol broadband dielectric spectrometer. Disc samples, 20 mm in diameter and 1 mm thick, were tested in frequency ranges between 10^6 and 10^{-1} Hz.

2.3 Sample preparation

The Sylgard 184 silicone kit was mixed in a 10:1 ratio by weight, as recommended by the manufacturer. Subsequently the desired amount of glycerol was added to PDMS and stirred with the help of the speed-mixer for 5 min at 3500 rpm, unless mentioned otherwise (3500 rpm is the maximum rotational speed of the device). No additional degassing of the formulations was necessary, due to the dual asymmetric method of mixing, which effectively pushes out entrapped air voids. After the mixing step, all compositions were cast onto a metal mould with a 1 mm spacer and cured at 80 °C for 1 h. Obtained films were then left at room temperature for at least two days, in order for eventual post-curing to take place. Samples for tensile tests were prepared according to ASTM D412-C standards. Mixed formulations were cast onto a PMMA plate with a 3 mm spacer. After curing, the samples were cut with a custom-made die.

The abbreviation 'phr', used to describe glycerol content in all compositions, corresponds to glycerol weight amount per hundred weight parts of silicone rubber. Sample names were formed using the pattern GX_S184, where G and X stand for glycerol and glycerol phr added to a PDMS prepolymer, respectively. Intuitively S184 corresponds to the applied PDMS system.

3 Results and discussion

3.1 Glycerol in PDMS emulsions and the morphology of cured samples

The morphology of glycerol in PDMS emulsions obtained by direct mixing the two virtually immiscible liquids was analysed with the help of optical microscopy. Proper mixing conditions were determined with a simple test, in which a composition of pre-polymer and glycerol was mixed at different speeds and increasing mixing times. It was found that an increase in shear forces has a tremendous impact on glycerol droplet size (see Fig. 1), in that the higher the rotational speed, the more mono-dispersed emulsions are obtained. The maximum droplet diameter decreased from around 30 µm to around 3 µm for the same formulations mixed for 5 min at 1000 and 3500 rpm, respectively. Longer mixing times were proved to have a negligible impact on droplet size, though for each glycerol concentration a minimum mixing time that ensures obtaining an agglomeration-free emulsion is required. Therefore, most compositions were mixed for 5 min at 3500 rpm, which provided sufficient shear forces for obtaining reasonably mono-dispersed emulsions for a broad range of glycerol incorporated into PDMS. The images shown in Fig. 2 present various uncured compositions with increasing amounts of incorporated glycerol. Viscosities of the obtained emulsions are presented in Fig. 3. Produced mixtures were stable for extensive amounts of time. No change in droplet size was observed, even several hours after mixing.

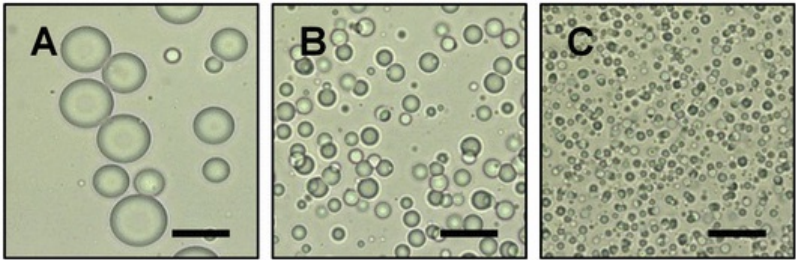


Fig. 1 Optical microscopy images of 10 phr glycerol in S184 emulsions obtained after 5 min of speed-mixing at 1000 (A), 2000 (B) and 3500 (C) rpm. Scale bars for all images correspond to 25 μm .

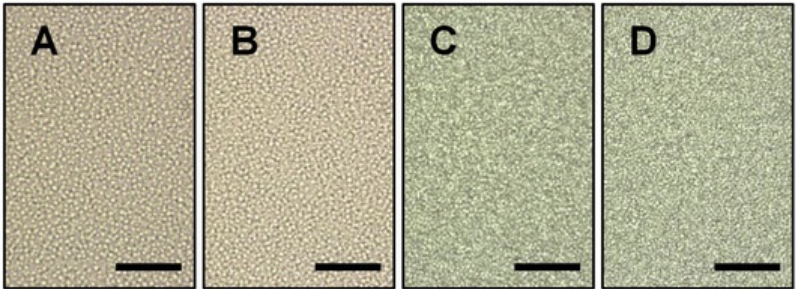


Fig. 2 Optical microscopy images of glycerol in PDMS emulsions. A – 10 phr, B – 40 phr, C – 70 phr, D – 100 phr. Scale bars for all images correspond to 50 μm .

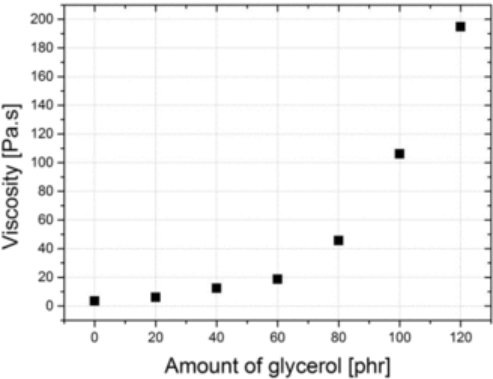


Fig. 3 Viscosities of various glycerol-Sylgard 184 formulations.

Cross-sections of crosslinked composites were investigated through scanning electron microscopy. The images presented in [Fig. 4](#) show the cross-sections of four samples with 10, 50, 90 and 130 phr of incorporated glycerol, respectively. As can be seen, when the amount of glycerol increases, droplet concentration becomes higher; however, the average droplet size remains almost uninfluenced. It was additionally observed that while increasing the glycerol/PDMS ratio of formulations, a threshold concentration is finally reached above which a droplet-like morphology is no longer present in the crosslinked material. Instead, a network with interconnected glycerol channels is formed. This phenomenon was additionally proved by testing the conductivity of the compositions. Broadband dielectric spectroscopy tests showed that the sample with 140 phr of glycerol exhibited slightly increased conductivity compared to formulations with lower glycerol concentrations (see [Fig. 5](#)). This is in agreement with our expectations, since it is common knowledge that glycerol exhibits higher electrical conductivity than PDMS. Therefore, if we consider that glycerol molecules are no longer encapsulated by the PDMS matrix, it can be expected that higher conductivity paths will be formed throughout the material. This will lead finally to an overall conductivity increase in the material. Glycerol conductivity is additionally enhanced by the unavoidable presence of impurities such as ion-containing water. This increased conductivity is considered as a direct indication of the formation of a bicontinuous phase in the elastomer.

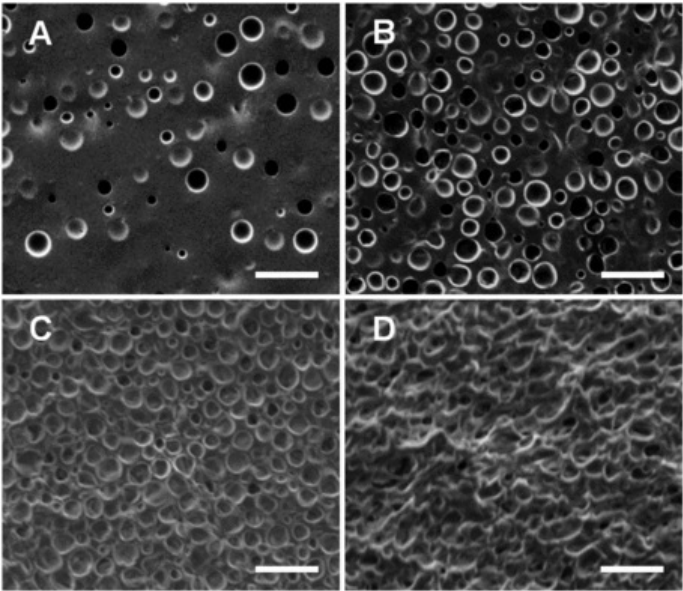


Fig. 4 SEM images of cured glycerol-PDMS composite cross-sections. A – 10 phr, B – 50 phr, C – 90 phr, D – 130 phr of glycerol. Scale bars for all images correspond to 10 μm .

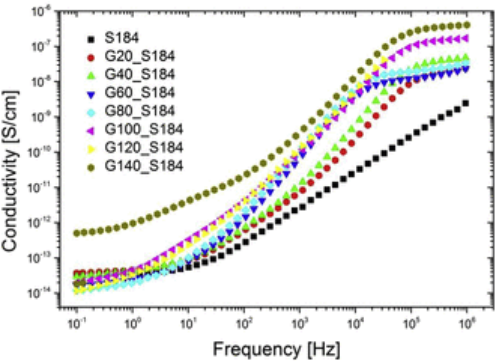


Fig. 5 Conductivity of various glycerol-PDMS compositions presented as a function of frequency.

Different compositions were additionally investigated by means of infrared spectroscopy. Cross-sections of 1 mm thick samples, containing 20, 60, 100 and 140 phr of glycerol, were prepared and tested with the ATR-FTIR technique. Pure Sylgard 184 was tested for reference. The obtained spectra are presented in Fig. 6 below. A broad peak typical for the O–H band at a wavelength of around 3400 cm^{-1} becomes more prominent for compositions with increasing amounts of glycerol. This again indicates the high efficiency of glycerol encapsulation within the PDMS matrix and proves that glycerol remains in the sample.

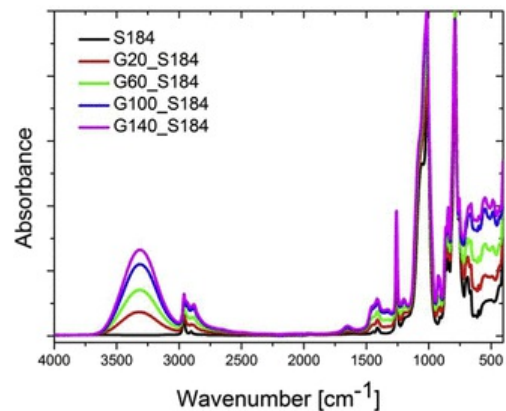


Fig. 6 ATR-FTIR spectra of various glycerol-PDMS composite cross-sections.

3.2 Water absorption

It was additionally observed that glycerol-PDMS composites exhibit an extraordinary ability to absorb water. Therefore, a series of simple experiments was conducted, in which disc samples (25 mm in diameter and 1 mm thick) were immersed in deionised water for 24 h and subsequently vacuum-dried. The mass, diameter and thickness of the samples were monitored before the experiment, after water absorption and after 1 and 3 days of drying. Results for the water absorption and drying experiments are presented in Fig. 7 and Fig. 8, respectively. It was found that samples with higher amounts of glycerol absorb water much more effectively than compositions with lower amounts – as expected. Increases in mass, thickness and diameter became more prominent for samples with increasing glycerol content, reaching the maximum level at 100 phr of glycerol, due to the relatively high permeability of silicones combined with the hygroscopic nature of glycerol induced by the presence of three hydroxyl groups in each glycerol molecule. For a given glycerol content (140 phr), continuity in the structure of glycerol becomes evident as the glycerol starts to be washed out, which is proved by significant mass loss after immersion in water. This phenomenon can be explained by the fact that the continuous glycerol phase, which becomes more prominent due to an increasing number of interconnected glycerol voids in the structure, allows water to flush out glycerol from the material. As a result, water is not absorbed by glycerol-filled voids and consequently the sample's mass increases only slightly or even – as in the case of the highest concentration – decreases.

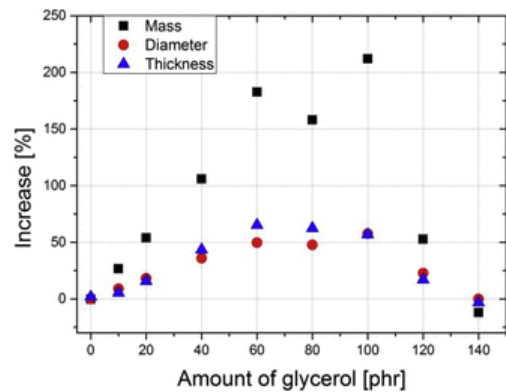


Fig. 7 Mass, diameter and thickness changes of various glycerol-PDMS formulations caused by water absorption.

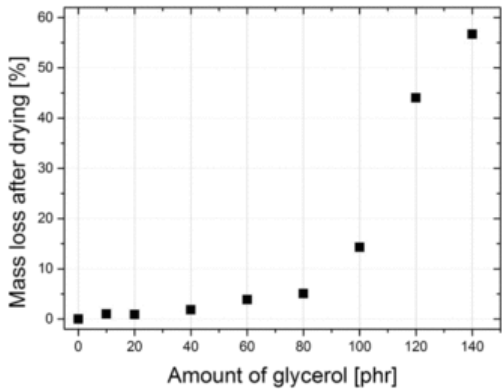


Fig. 8 Mass losses of various glycerol-PDMS formulations after 3 days' drying, resulting from water absorption experiments.

Furthermore, an increase in the stiffness of water-absorbed samples was observed. This stiffness gradually increased in line with increasing amounts of incorporated glycerol, caused by the developing stress as a result of expanding water-absorbing glycerol-filled voids in the material. Consequently, the developed stress induced changes in the shape of the samples. An example is presented in Fig. 9, where the sample with 40 phr of glycerol rolled after absorbing water, yet the original sample shape fully recovered after drying.



Fig. 9 Images of a 25 mm in diameter and 1 mm thick sample, G40_S184, before the absorption experiment, after absorption and after drying, respectively.

3.3 Dried materials

Ideally, after drying, elastomer should recover to its initial size, due to water being released from the material. Nevertheless, a small decrease in mass was observed for samples with ≤ 80 phr of glycerol. In these cases glycerol was partially flushed out from the material and replaced by water. During the drying process water left the voids, which finally led to a mass decrease in the sample. Nevertheless, in the case of samples with ≥ 100 phr, the mass decrease became substantial. Since, as it has been hypothesized, glycerol voids are interconnected in the presence of high glycerol content, it was therefore much easier for water to flush out glycerol from the material. It finally led to a mass decrease of 14%, 45% and 57% with respect to initial sample mass for the samples with 100, 120 and 140 phr of glycerol, respectively. Mass sample losses are presented in Fig. 8, in which it is evident that the mass losses become more prominent in line with increasing glycerol content. Nevertheless, the difference between the samples with 80 and 100 phr of glycerol is more significant, whereas samples with 120 and 140 phr exhibit major mass losses, thereby indicating that the vast majority of glycerol was flushed away from the samples. Based on these experiments we believe that glycerol-PDMS composites can be applied, for example, as a tool for controlled drug delivery, where adjustable release velocity is required.

3.4 Tensile tests

Samples with various amounts of glycerol incorporated into PDMS were tested. The mechanical behaviour of Sylgard 184 was investigated for reference purposes. Five specimens from each composition were tested, and average values were calculated and used for evaluating the mechanical properties of the material. Values of tensile modulus of elasticity as well as ultimate stresses and strains are listed in Table 1, and averaged curves for each tested composition are presented in Fig. 10 below.

Sample	Tensile modulus of elasticity [MPa]		Strain at break [%]	Stress at break [MPa]
	Tangent (Y)	Secant (Y_{100})		
S184	1.07	3.66	154	8.29

G20_S184	0.80	2.58	160	6.19
G40_S184	0.58	1.76	166	4.60
G60_S184	0.51	1.55	168	4.04
G80_S184	0.43	1.32	161	3.17
G100_S184	0.38	1.14	159	2.71
G120_S184	0.33	0.97	116	1.30

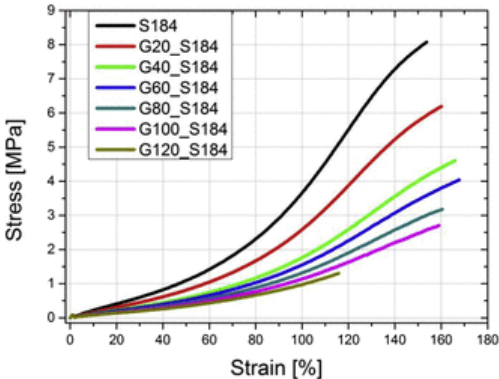


Fig. 10 Stress-strain behaviour of various compositions obtained from tensile measurements.

From the curves in Fig. 10 it can be seen clearly that stress produced on samples through mechanical displacement decreases in line with increasing amounts of incorporated glycerol. All compositions exhibit typical linear elastic behaviour up to strain values of around 40%, as reported also by other groups investigating PDMS systems [23,24]. The ultimate stress as a function of glycerol content is presented in Fig. 11. Pure Sylgard 184 exhibits the highest stress, which dramatically decreases when incorporating 20 phr of glycerol, while incorporating 40 phr of glycerol decreases the stress even further. Nevertheless, an additional increase in glycerol content influences stress less significantly but preserves the same tendency as in the case of lower glycerol loadings. In general the decrease in stress is attributed to the presence of glycerol droplets in the material's structure, which introduces zones of zero stress and finally leads to an overall decrease in stress. On the other hand, it is believed that the glycerol voids, to some extent, can absorb tension produced while stretching the material and act as a plasticiser by partially decreasing friction in the material. Therefore a sharp decrease in mechanical strength and a slight increase in maximum strain are observed. Glycerol has no inhibiting effect on the platinum catalyzed crosslinking reaction and is thus not supposed to alter the crosslinking density. Moreover it has been reported that PDMS does not swell in the presence of glycerol [25]. This fact is additionally supported by comparing solubility parameters, which are $\delta = 7.3 \text{ cal}^{1/2} \text{ cm}^{-3/2}$ and $\delta = 21.1 \text{ cal}^{1/2} \text{ cm}^{-3/2}$ for PDMS and glycerol, respectively [25]. We consider this as a direct indication that glycerol does not interfere chemically or physically during the crosslinking process and that ultimate composite properties are not influenced thereupon.

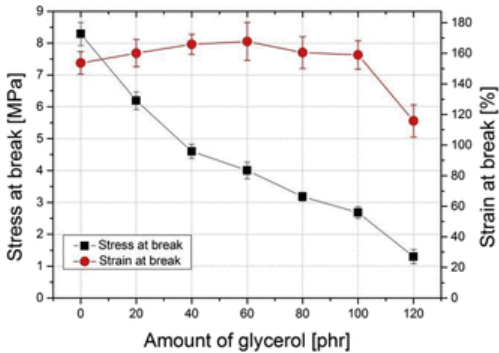


Fig. 11 Ultimate properties of various glycerol-S184 compositions.

From Fig. 11 it is evident that the ultimate strain increases in line with increasing glycerol loading and reaches its maximum at 60 phr of glycerol in PDMS. This phenomenon can be attributed to the fact that the stress exerted on samples can partially be overcome by glycerol voids, which most likely absorb some of the tension. A further increase in the amount of glycerol decreases maximum strain, which in turn drops drastically after exceeding 100 phr of glycerol. The results suggest that incorporating ≥ 80 phr of glycerol negatively influences material integrity, which is not significant for samples with 80 and 100 phr of glycerol but becomes prominent at 120 phr of glycerol.

The tensional tangent modulus of elasticity (Y) was calculated from a slope of averaged stress-strain curves, while the tensional secant modulus of elasticity (Y_{100}) was calculated from a stress/strain quotient at 100% strain. All calculated values are summarised in Fig. 12. It can be seen clearly that the values of both moduli decrease in line with increasing glycerol loading in Sylgard 184. The most prominent loss can be observed again for samples in the range 0 phr–40 phr of glycerol, whereas changes in moduli at higher loadings are less substantial, which is in agreement with previous observations.

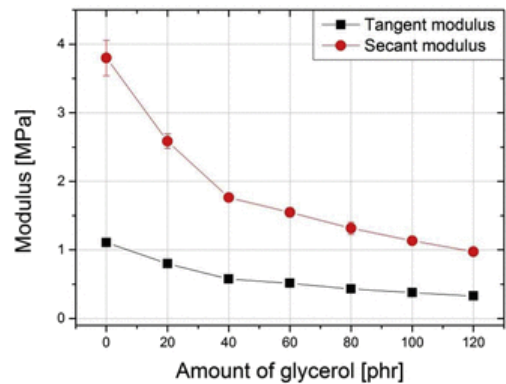


Fig. 12 Tensional tangent (Y) and secant (Y_{100}) moduli of elasticity of various glycerol-S184 compositions.

Optical inspection of the stretched material was performed on 50 μm thick PDMS films containing 5 phr of glycerol. The samples were stretched up to 100% and 150% strain values, in order to investigate the behaviour of glycerol-filled voids under applied mechanical stress. An optical microscopy image presented in Fig. 13-A shows that glycerol beads present in the sample and stretched by 100% become elongated in the stretch direction. Glycerol droplets remain stable at this point and do not interconnect with each other. Nevertheless, when the 150% strain is applied, the stress produced on the elastomer forces the glycerol to leave the voids. This phenomenon is depicted in Fig. 13-B, where small glycerol droplets can be observed on the sample's surface. This unique behaviour indicates that the composite, to some extent, loses its integrity at very high stretch ratios. This phenomenon is considered as a materials feature that may potentially lead to various applications where stress-induced substance release is required.

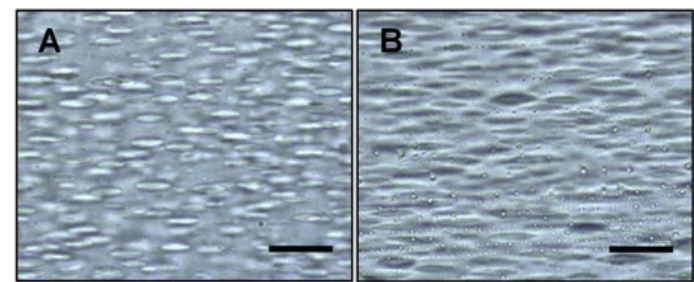


Fig. 13 Optical microscopy images of stretched glycerol-PDMS composites (5 phr). A – 100% strain, B – 150% strain. Scale bars for all images correspond to 25 μm .

4 Conclusions

An innovative type of elastomeric material was prepared and tested. In this approach a counterintuitive mixture of hydrophilic glycerol and hydrophobic PDMS was prepared as the basis for a green and cheap elastomer. The obtained glycerol-in-PDMS emulsion was very stable in a broad range of concentrations, even while processing the pre-polymer formulations. The ultimate strain of the elastomer was not compromised by incorporating glycerol, even at higher loadings. Furthermore, material properties like Young's modulus can be modified easily by altering the amount of incorporated glycerol. Additionally, the price of our material is significantly cheaper compared to pure PDMS, due to negligible costs of glycerol as a by-product in biodiesel production. Considering that the glycerol contents described herein substantially exceed 100% by weight, the material's overall price can be reduced at least twofold.

Apart its counterintuitive mechanical properties, the investigated composition exhibits very particular behavior in the presence of water. Water is absorbed by the material, and additionally glycerol (and potentially other substances) is released from its structure. From the perspective of designing smart materials, this invention could be of a great importance, for example, in the field of controlled drug delivery, water absorbing elastomers, sealants and even soft machines [26]. Furthermore the material may be designed into functional patches with applications for wound care or vehicles for skin therapeutics [27,28]. Based on our discussion, it can be assumed that by modifying the PDMS matrix, glycerol-PDMS composites can exhibit different release velocities which can potentially lead to broadening the application spectrum.

Acknowledgements

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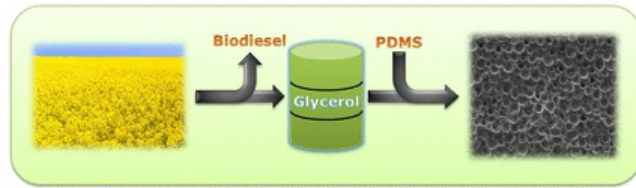
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Graphical abstract



Highlights

- A novel two-phase glycerol-silicone elastomer is presented.
- Glycerol is incorporated into PDMS in a form of discrete droplets.
- The resulting composite can be considered as ecological and inexpensive.
- The inclusion does not affect significantly the ultimate strain of the elastomer.
- The composites exhibit very particular behaviour in the presence of water.

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